Molten Salt Reactor
PROGRAM

Development of MSR Modeling Capabilities for Solubility Prediction

Kyoung Lee, Bob Salko, Joanna McFarlane

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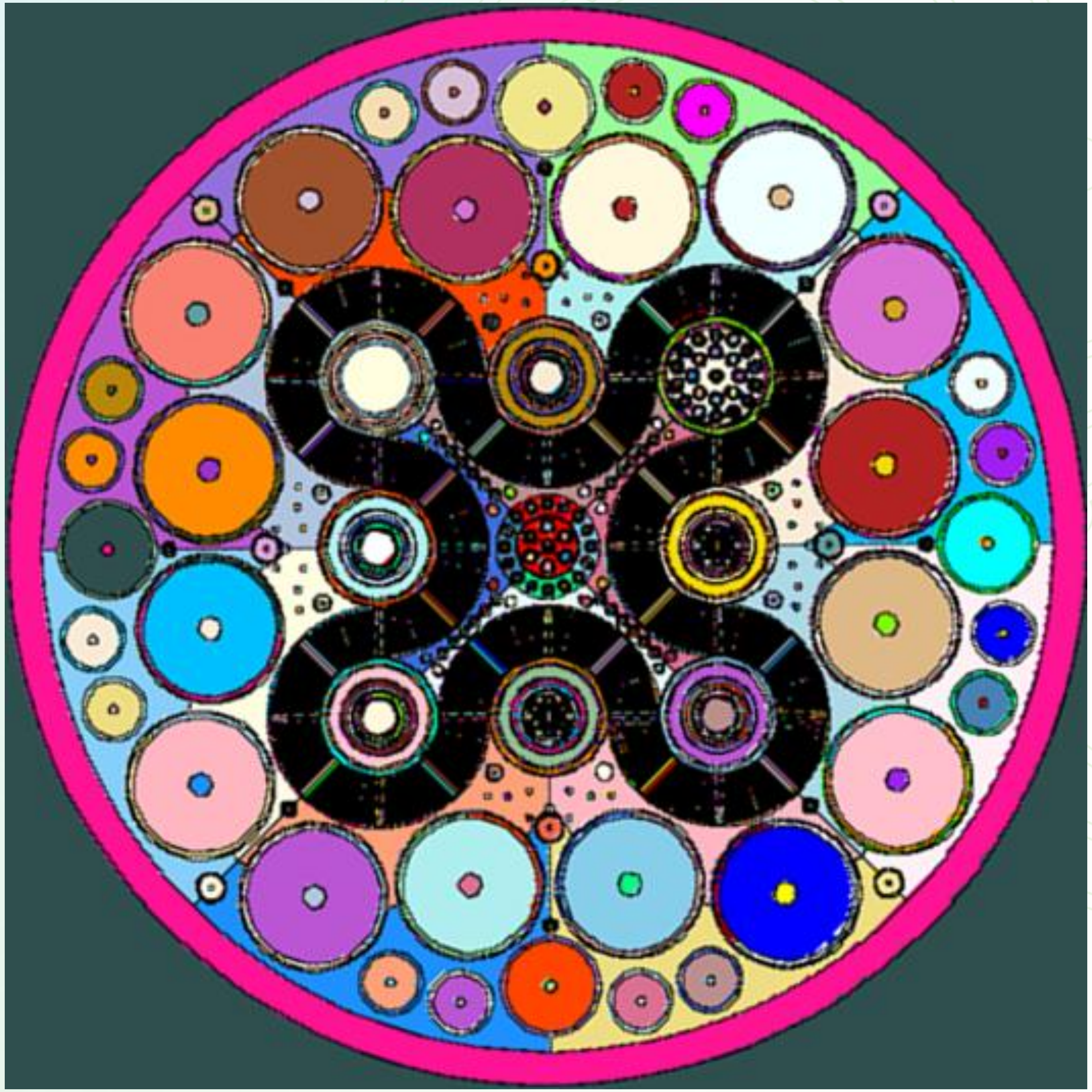
Overviews

Calculate the depletion of a salt target in the ATR using the Oak Ridge Isotope GENeration (ORIGEN) code. The salt target composition is FLiBe ($2LiF\text{-}BeF_2$).

Determine the elemental composition derived from fission products. The primary motivation for irradiation is to produce these fission products from the splitting of heavy atomic nuclei like uranium-235.

Develop MSR (Molten Salt Reactor) and gas phase compositions using Thermochimica (or FactSage).



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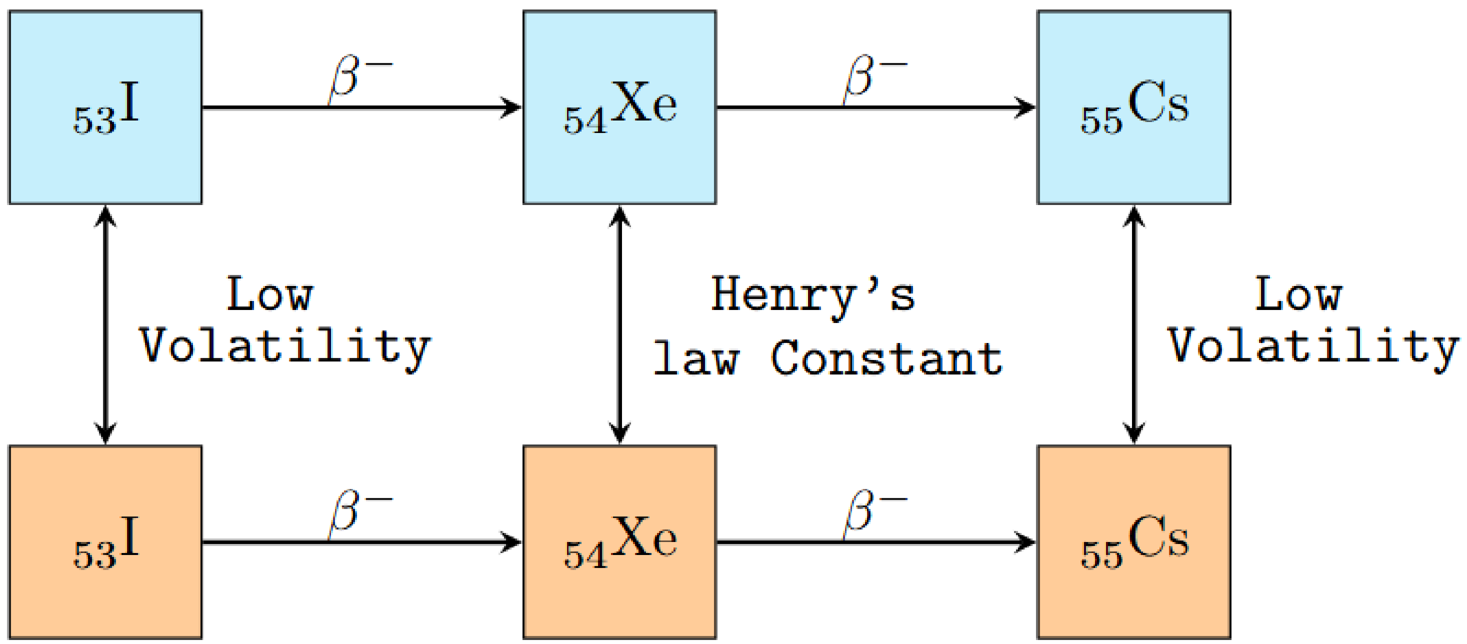
Using Gibbs Free Energy Minimizer to Model Low Volatility Gas Behavior

Introduce a framework to predict gas phase behavior and solubility using temperature, pressure, and composition inputs.

Gibbs Energy Minimizer (GEM): Used to predict equilibrium concentrations and phase distributions in complex mixtures, particularly for low-volatility gases.

Methodology:

1. Apply Gibbs energy minimization to estimate iodine solubility in molten salts.
2. Use FactSage and Thermochimica for Gibbs energy-based calculations.
3. Include temperature, pressure, and mole fraction dependencies in the model.



Optimize Gibbs free energy calculations to improve iodine and cesium gas behavior predictions over a wide temperature range.

Thermodynamic Models:

- **Henry's Law**: Describes gas solubility in liquids.
- **Raoult's Law**: Governs vapor pressure of solvents in solutions.

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Henry's law constant

Henry's law constant describes dissolved gas proportionality in liquid-gas equilibrium.

- Noble gas partitioning affects MSR performance.
- Liquid-gas mass transport in determining reactor parameters.
- Gibbs free energy essential for understanding phase transitions.

Gibbs free energy $\Delta G = \Delta G_\gamma + \Delta G_v$

$$\Delta G(r, T; \gamma(T), \alpha, \beta) = RT \ln(K_H) = 4\pi r^2 \alpha \gamma(T) + \frac{4}{3}\pi r^3 \beta RT,$$

surface tension

$$\gamma(T) = \frac{\partial \gamma(T)}{\partial T}(T - 273.15) + \gamma_0.$$

enthalpy change

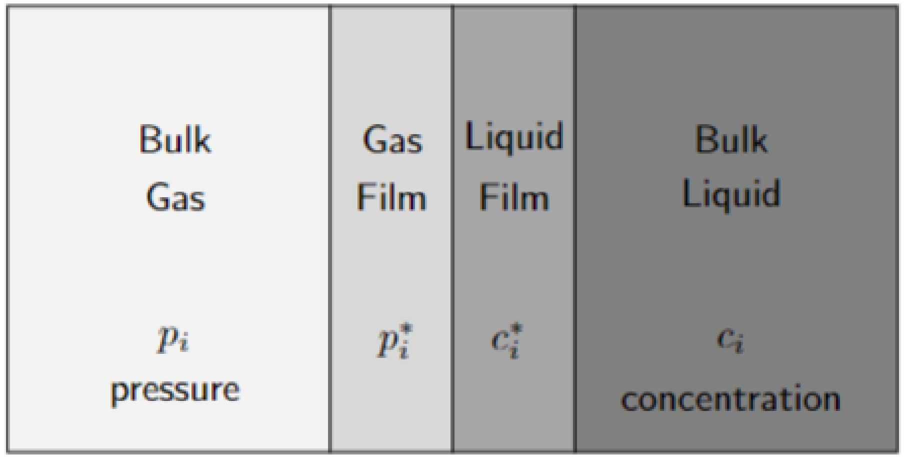
$$\Delta H = RT^2 \frac{d \ln(K_H)}{dT} = -4\pi\alpha r^2 \gamma_0$$

entropy change

$$\Delta S = -8\pi\alpha r^2 \frac{\gamma(T)}{T} + 4\pi\alpha r^2 \frac{\partial \gamma(T)}{\partial T} - \frac{4}{3}\pi r^3 \beta R.$$

- **Henry's Law** is used to describe the solubility of gases in liquids, where the gas does not chemically react with the solvent. – high volatility.

$$A(g) \rightleftharpoons A(l)$$



$$c_i^* = p_i H$$

$$p_i^* = c_i / H$$

The understanding of the gas–liquid interface is clarified by the two-film theory, which elucidates this phenomenon by utilizing partial pressures and concentrations, where H represents Henry's law constant.

Lee, K.O., Williams, W.C., McFarlane, J. *et al.* Semi-empirical model for Henry's law constant of noble gases in molten salts. *Sci Rep* **14**, 12847 (2024). https://doi.org/10.1038/s41598-024-60006-9

$$[\mathrm{I}]_{input} = [\mathrm{I}]_c(l) + [\mathrm{I}]_c(g)$$

$$k_g = \frac{[\mathrm{I}]_c(g)}{[\mathrm{I}]_{input}}$$

$$k_l = \frac{[\mathrm{I}]_c(l)}{[\mathrm{I}]_{input}}$$

$$k_s = \frac{[\mathrm{I}]_c(l)}{[\mathrm{I}]_c(g)}$$

$$[\mathrm{I}]_{input} = [\mathrm{I}]_c\,(l) + [\mathrm{I}]_c\,(g)$$

$$[\mathrm{I}]_c\,(g) = [\mathrm{I}]\,(g) + [\mathrm{I}_2]\,(g) + \cdots$$

$$\mathrm{I}_2\,(g) \rightleftharpoons 2\mathrm{I}\,(g)$$ decomposes at high temperature, endothermic

$$2\mathrm{I}^-\,(g) \rightleftharpoons \mathrm{I}_2\,(g) + 2e$$ To convert iodide ions to be oxidized to iodine gas

FLiBe, 2LiF-BeF_2

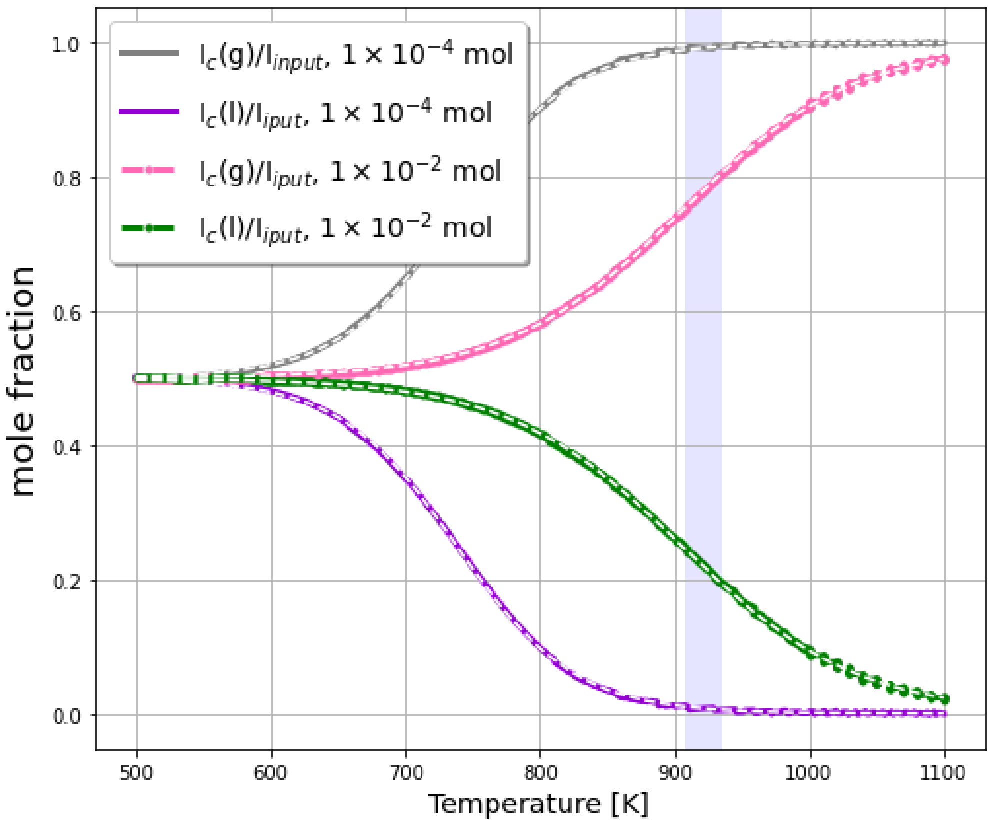
$$s(x,i) = l(x,i)/g(x,i)$$

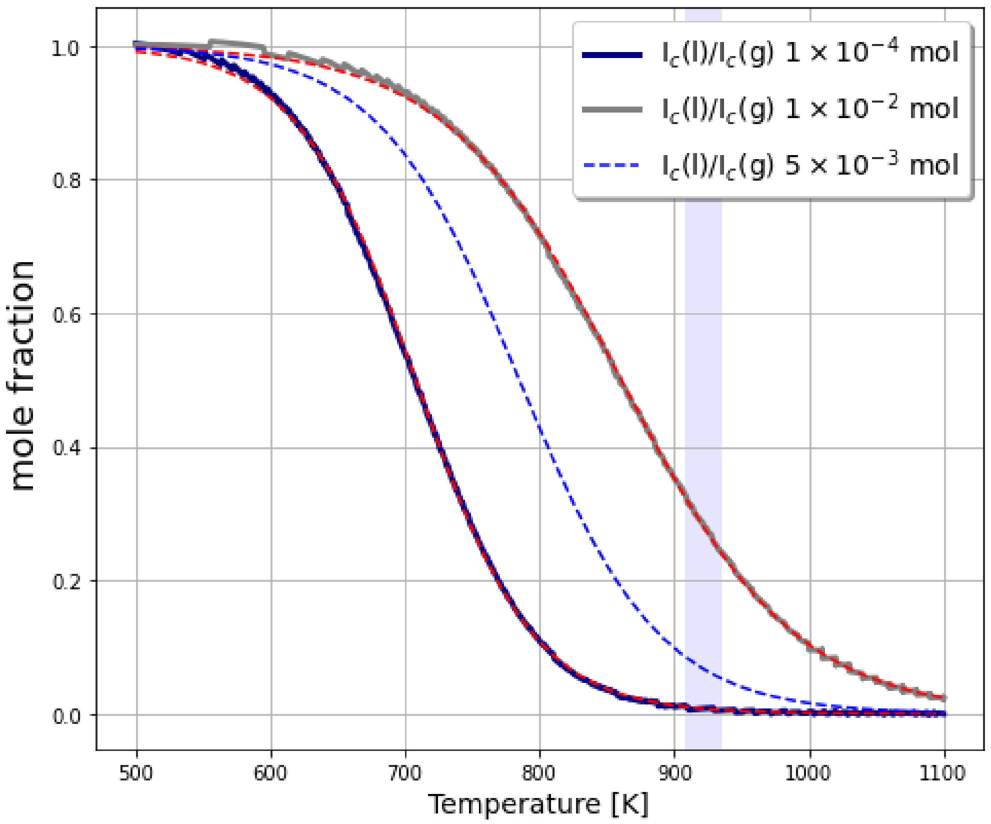
$$g(x,i) = \frac{a}{1+\exp\left[-k_i(x-T_i)\right]} + a$$

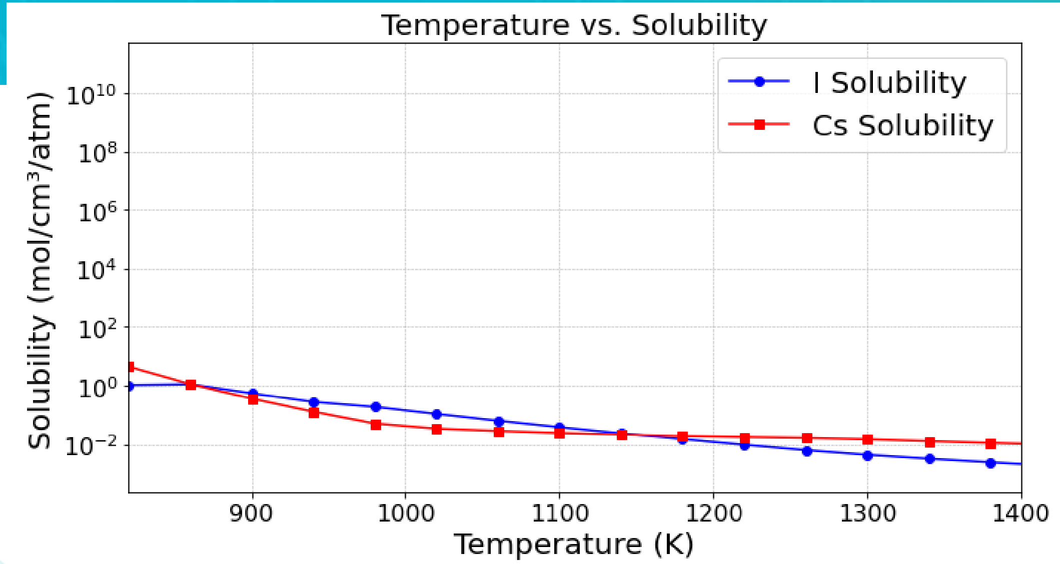
$$l(x,i) = \frac{a}{1+\exp\left[k_i(x-T_i)\right]}$$

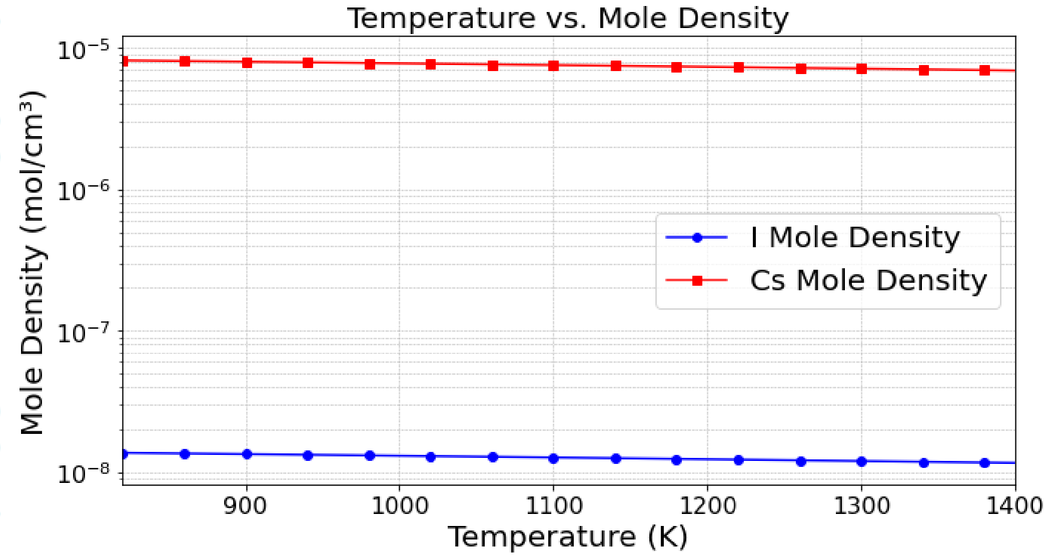
$$k_i = k_0 + k_1 i$$

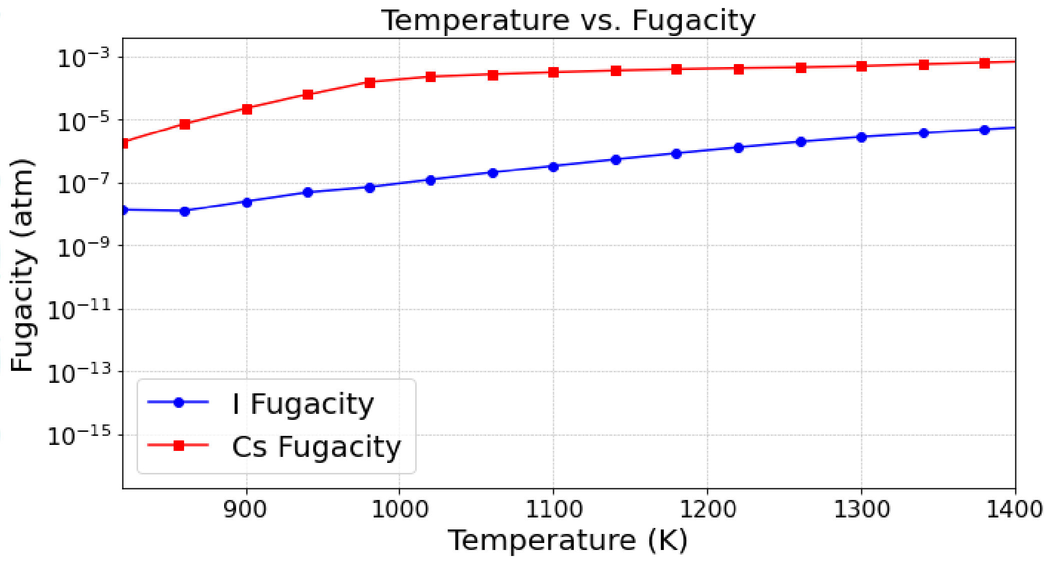
$$T_i = T_0 + T_1 i$$

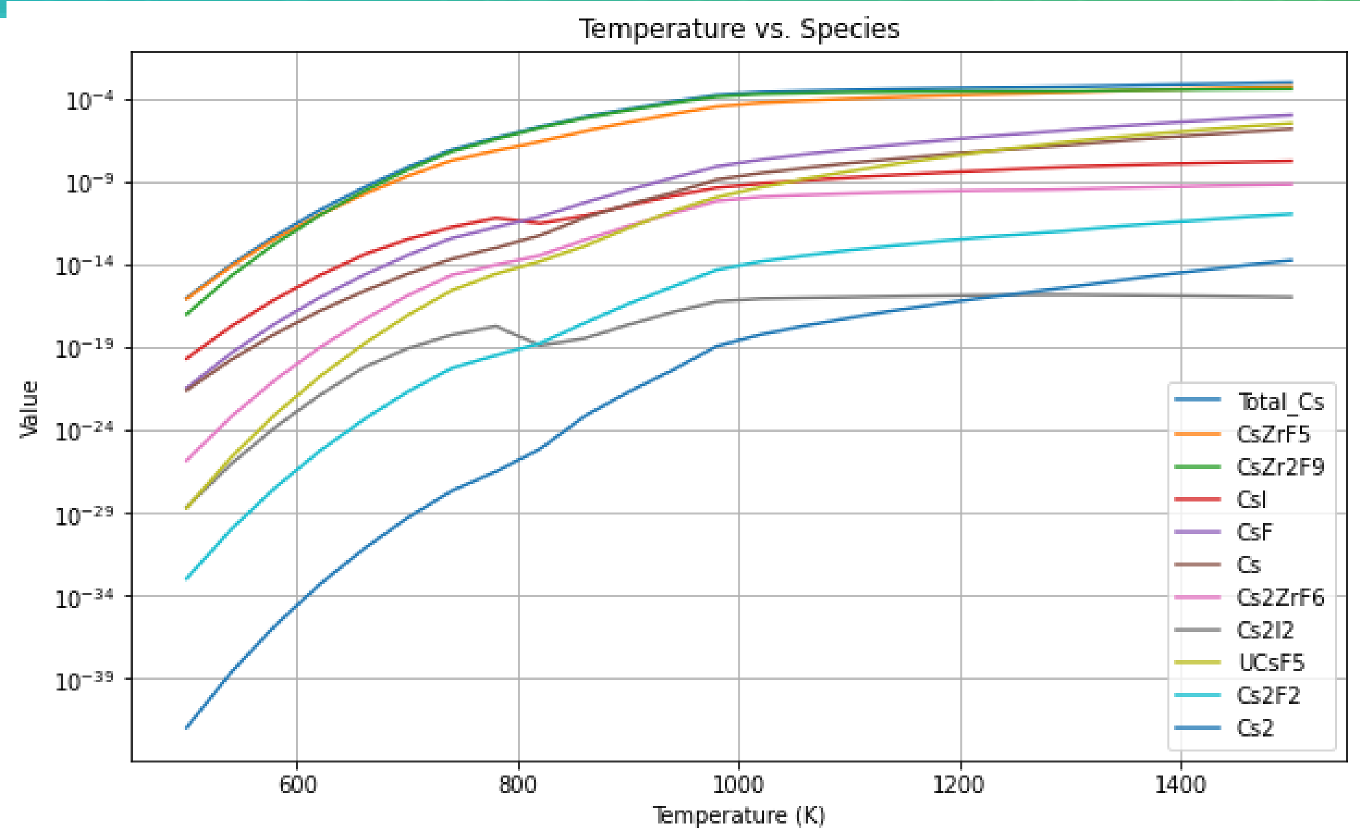


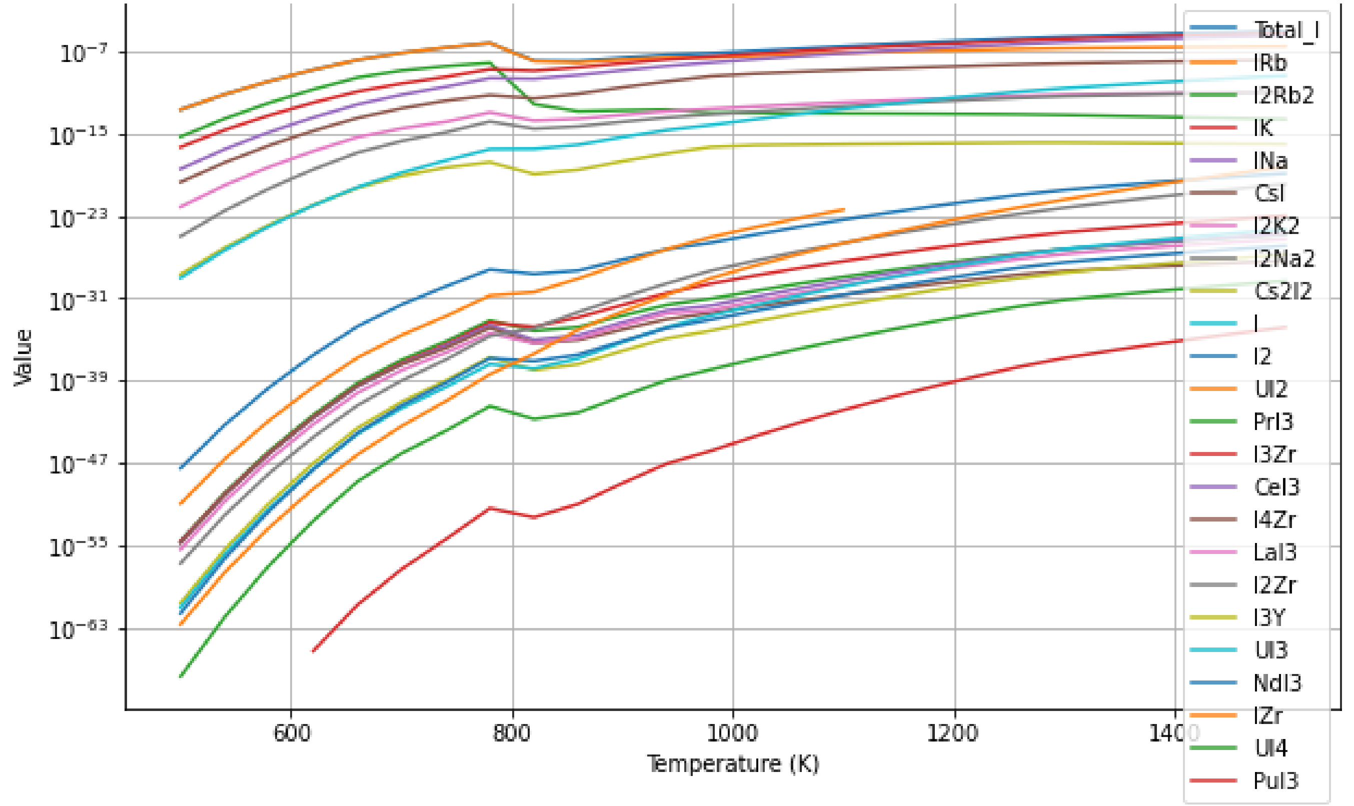



Temperature vs. Solubility
Solubility (mol/cm³/atm)
Temperature (K)
I Solubility
Cs Solubility


Temperature vs. Mole Density
Mole Density (mol/cm³)
Temperature (K)
I Mole Density
Cs Mole Density


Temperature vs. Fugacity
Fugacity (atm)
Temperature (K)
I Fugacity
Cs Fugacity


Temperature vs. Species
Value
Temperature (K)
Total_Cs
CsZrF5
CsZr2F9
CsI
CsF
Cs
Cs2ZrF6
Cs2I2
UCsF5
Cs2F2
Cs2


Value
Temperature (K)
Total_I
IRb
I2Rb2
IK
INa
CsI
I2K2
I2Na2
Cs2I2
I
I2
UI2
PrI3
I3Zr
CeI3
I4Zr
LaI3
I2Zr
I3Y
UI3
NdI3
IZr
UI4
PuI3

OAK RIDGE National Laboratory

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Development of MSR modeling capabilities in SAM

Bob Salko, Kyoung Lee, Travis Mui, Rui Hu

U.S. DEPARTMENT OF ENERGY

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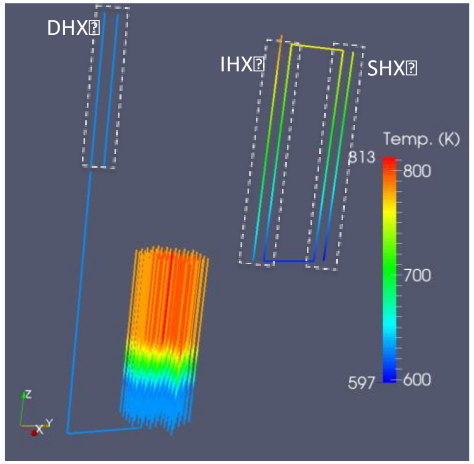
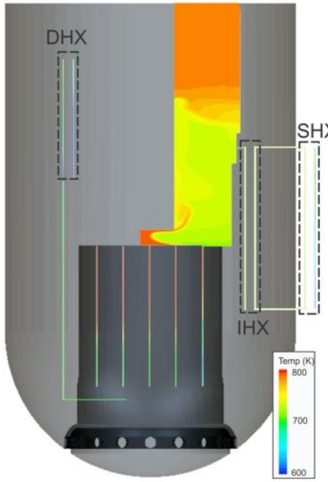
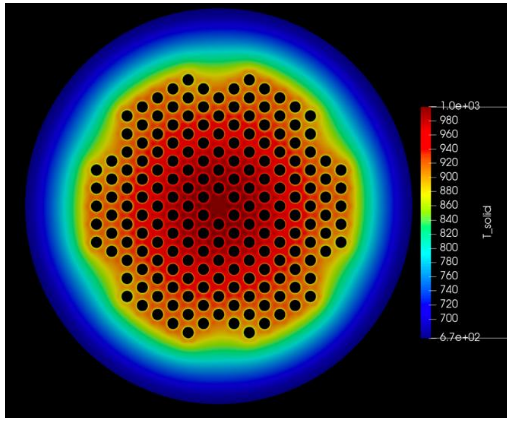
SAM Overview

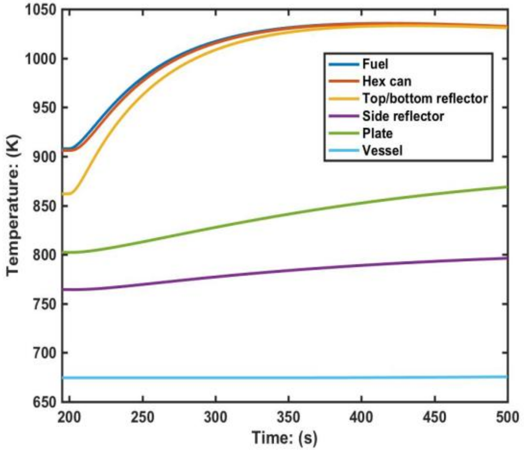
- A modern system analysis code for advanced non-LWR safety analysis.
  - Cover almost all non-LWR concepts.
- Advances in software engineering, numerical methods, and physical models (built-on MOOSE framework and its libraries);
- Advanced modeling features for various phenomena in advanced reactors;
- Flexible multi-scale multi-physics integration with other MOOSE- or non-MOOSE-based tools.
- 2019 R&D 100 Award.
- Being developed by NEAMS for MSR analysis by implementing species and gas transport features





Stand-alone and coupled SAM and CFD code simulations of SFR





Transient multi-physics simulation of heat-pipe-cooled micro-reactor

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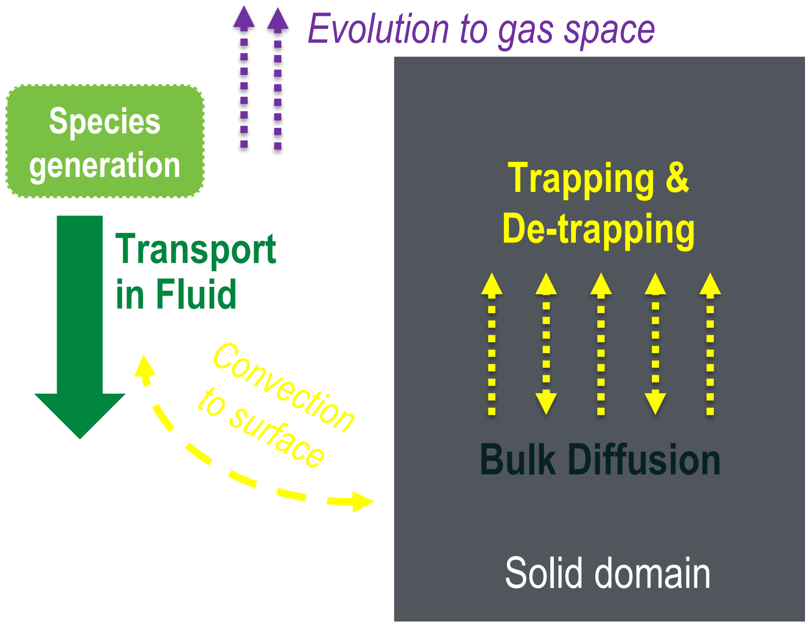
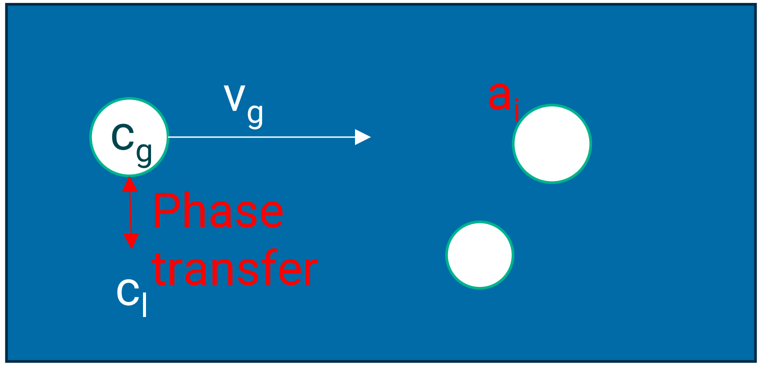
Species tracking in SAM

- Generic species (passive scalar) transport originally implemented in SAM to model flowing delayed neutron precursors in MSRs
- Framework expanded to simulate transport pathways of tritium in FHRs and MSRs
- Ongoing work to consider additional applications:
  - Oxidation-corrosion in lead-cooled reactors
  - Redox corrosion in salt-cooled reactors
  - Noble gas/metal transport in MSR



Species transport pathways modeled in SAM

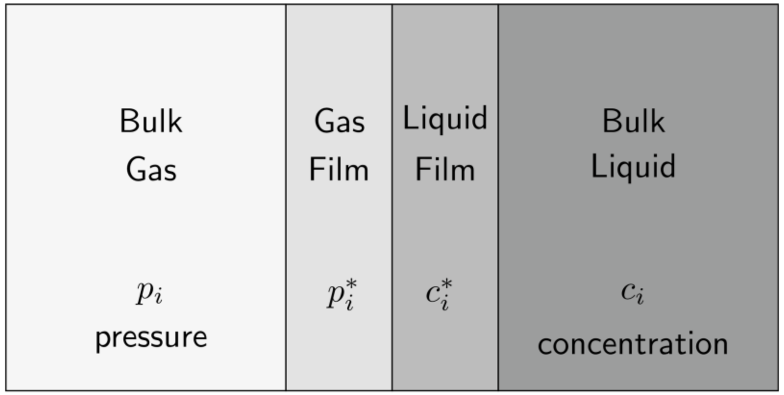
SAM predicts gas volume fraction, velocity, and bubble interfacial area



Species can exist in gas or liquid phases with phase transfer by Henry's Law

Coupling to Saline and Mole

- Mole provides modeling of high-volatile species phase transfer
  - Utilizes two-film theory
  - Provides Henry coefficients for FLiNaK and FLiBe salts and several noble gases
  - Mass transfer coefficient determined using Wilke-Chang or Stokes-Einstein[1]
- Saline is a C++/Fortran/Python interface to MSTDB-TP
  - Integrated into MOOSE and SAM
  - Provides thermophysical properties of many common salts and salt compositions



$$c_i^* = p_i H$$

$$p_i^* = c_i / H$$

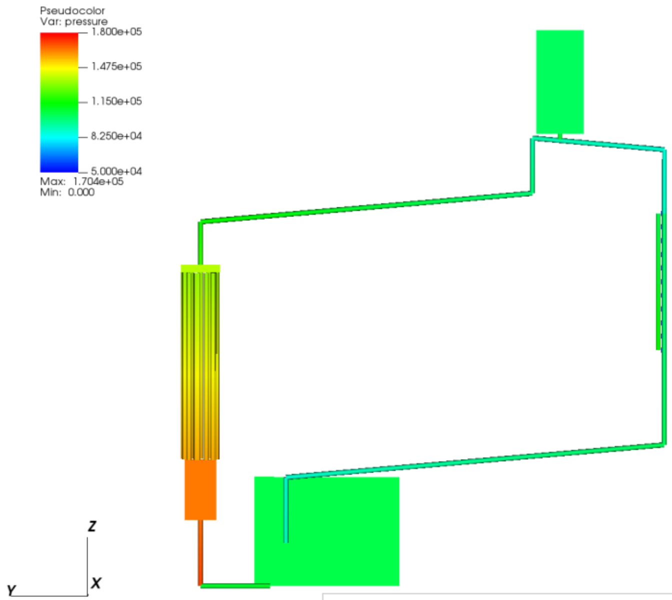
Gas liquid interface modeled using two-film theory

$$\Phi_j = K_j^L\left(p_j^g H_j - c_j^l\right) = K_j^G\left(p_j^g - c_j^l/H\right)$$

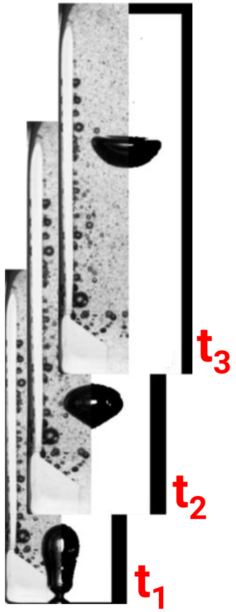
Interface concentrations determined using Henry's Law

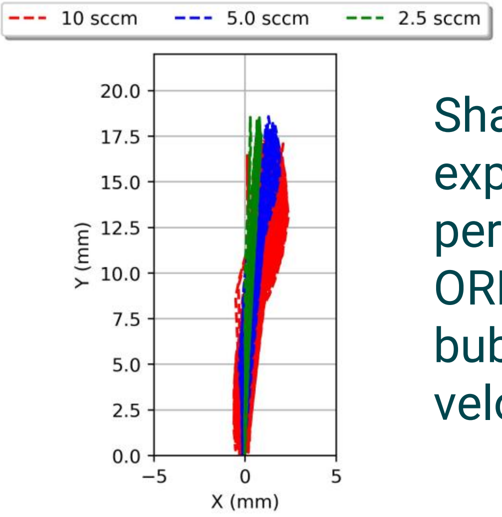
K. Lee, W. Williams, J. McFarlane, D. Kropaczek, and D. de Wet, "Semi-empirical model for Henry's Law Constant of Noble Gases in Molten Salts", Nature Portfolio, 14:12847, 2024, https://doi.org/10.1038/s41598-024-60006-9.

Modeling of LSTL and 10 ml salt experiments



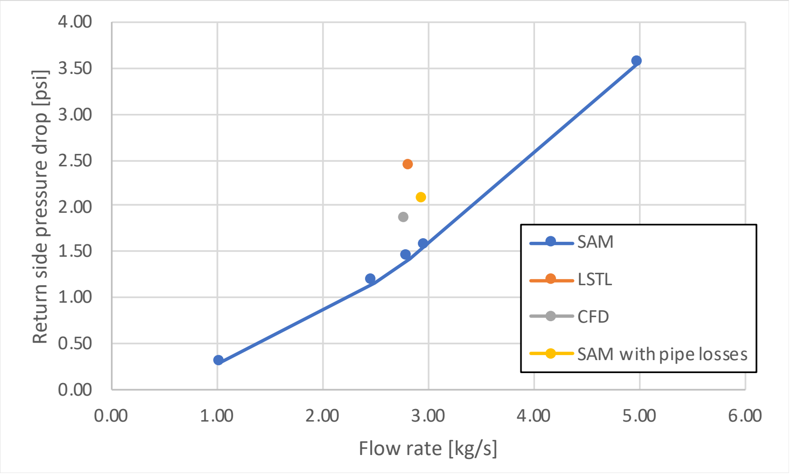
Pressure distribution predicted by SAM in LSTL

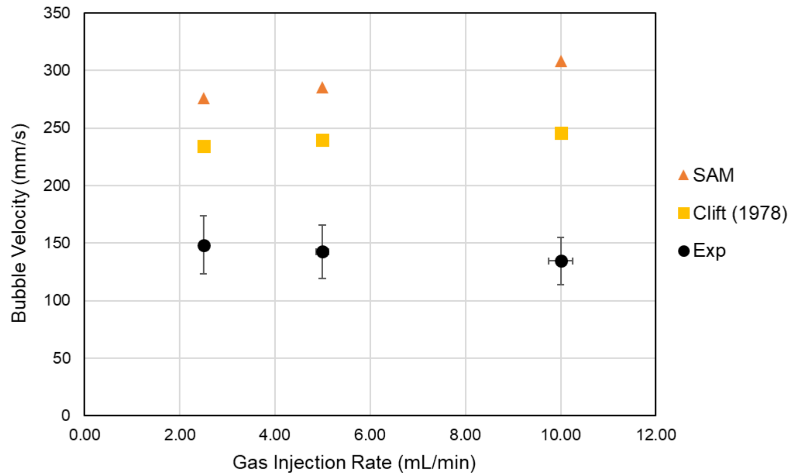




Shadowgraph experiments performed at ORNL to measure bubble terminal velocity in salt[1]

Comparison of SAM prediction (no form losses) to CFD and experimental results





Comparison of models and experiments show importance of wall effects and led to construction of new facility

1. McFarlane, J., et al. "Design of Instrumentation for Noble Gas Transport in LSTL Needed for Model Development" Report. ORNL TM-2023-3138 (2023).

2. Clift, Roland. "Bubbles." Drops and Particles (1978).

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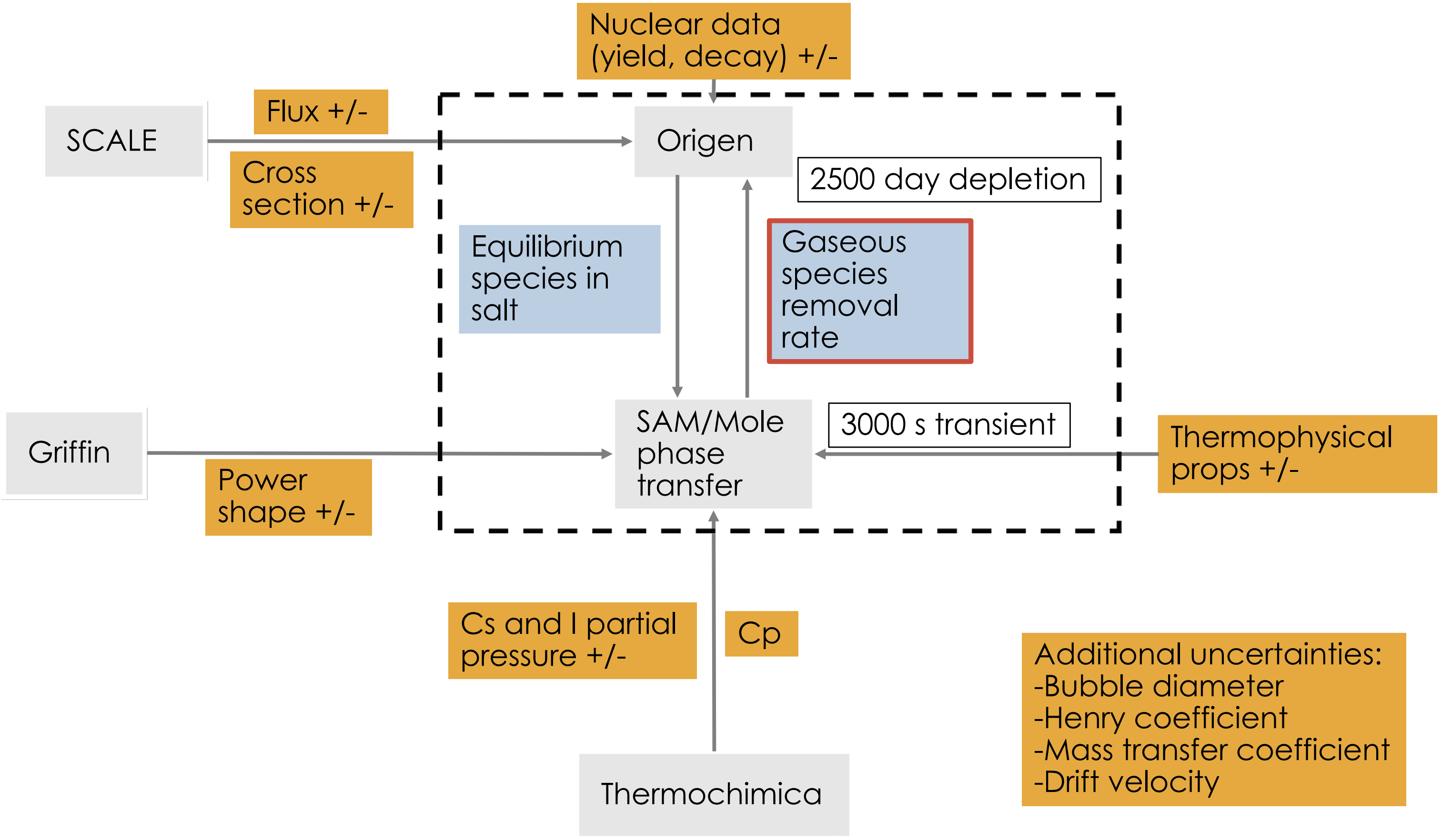
Modeling of off-gas system

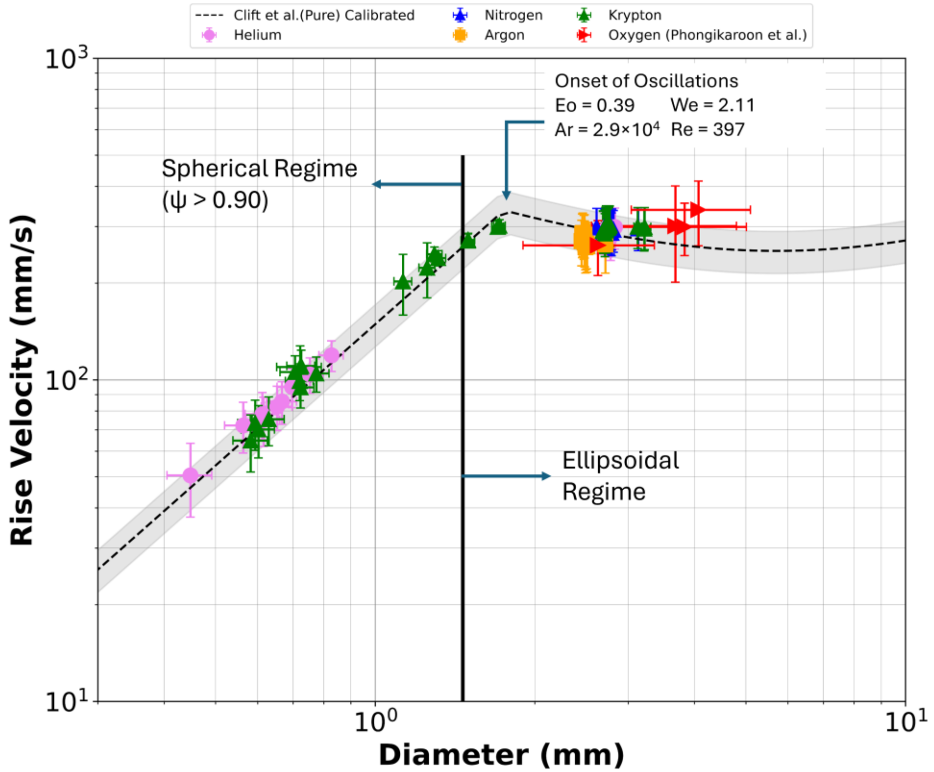
- Model pool-type MSR
  - Bubble generation due to xenon creation and phase transfer
  - Species and bubble transport affects fission gas concentration in pool region
  - Bubble buoyancy leads to escape at pool surface
- Depletion determines fission product inventory

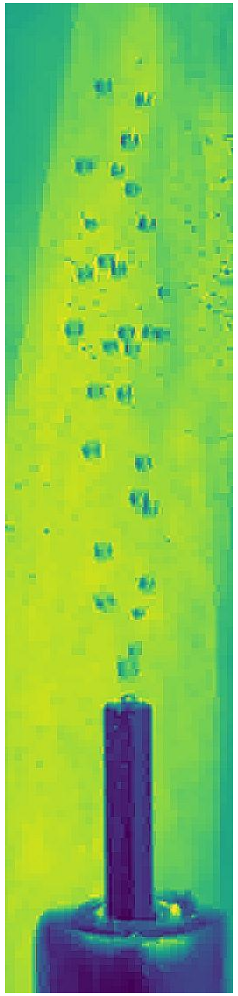


Coupled system being utilized for prediction of off-gas of fission products plus uncertainty

Uncertainty quantification design

- Forward UQ analysis
  - Variation of 15 input parameters
  - Nuclear data, thermophysical fluid properties, mass transfer data, bubble behavior
  - Observation of xenon, cesium, and iodine off-gas rate as FOMs
  - 5,000 evaluations of coupled analysis lead to convergence of FOMs
- Bubble velocity uncertainty obtained from MSR Campaign data





Molten salt bubble injection tests[1] used for quantification of terminal velocity uncertainty

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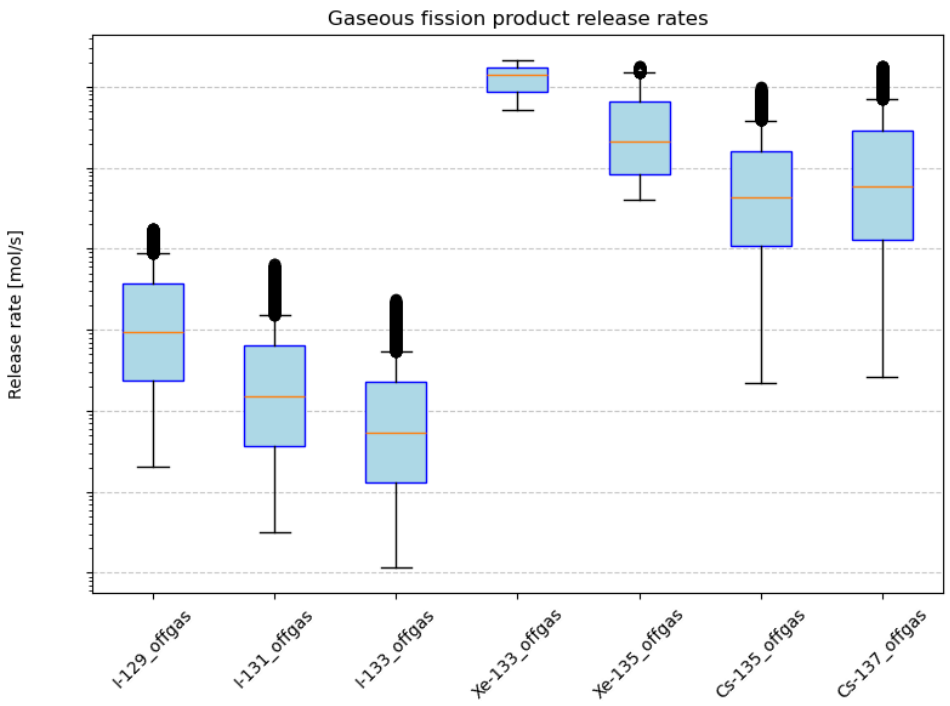
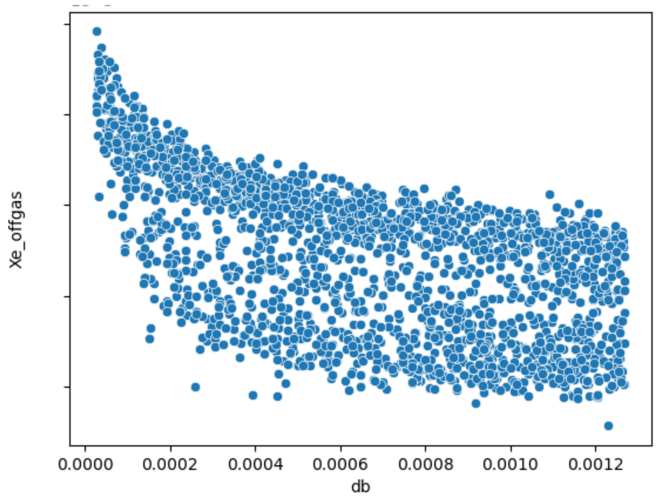
1. D. Orea, K. Robb, J. McFarlane, *Optical Measurements of Gas Bubble Rise Velocity in Molten LiCl-KCl*, in press., 2025

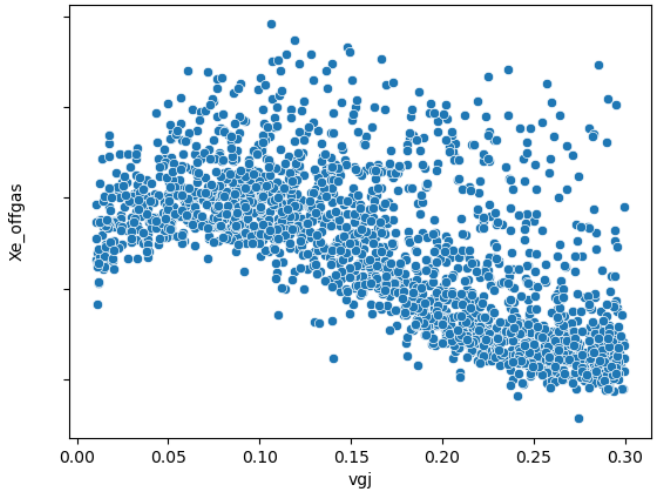
Prediction of fission gas release rates and uncertainty

- Determination of fission gas release rates plus uncertainty
- Bubble rise velocity is of high importance in determining fission gas release
- Bubble diameter is of high importance in determining interfacial area and phase transfer rate of high volatile
- Low-volatile partial pressures of high importance in determination of low-volatile release rates



Spread in offgas release rates due to input uncertainties





Relationship between input parameters and offgas rates

Summary

- Species and gas transport modeling capabilities expanded in SAM for modeling of MSRs
- Phase transfer modeling capabilities added through coupling of Mole
- Demonstration of capabilities shown by performing fission gas release uncertainty analysis
- Model will be expanded including coupling with Thermochimica, bubble modeling improvements based on MSR Campaign data, graphite xenon absorption, and improved coupling for depletion

Acknowledgements

- SAM/Mole development activities have been supported by the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program
- Demonstration of NEAMS tools for MSR UQ analysis has been funded by a US DOE iFOA project in collaboration with Terrestrial Energy, USA

Questions?

Development for MSR applications

- Fission gas behavior plays an important role in MSR operation
  - High volatile xenon gas is a neutron poison
  - Low volatile gases like cesium and iodine are source terms
- Prediction of gas location is needed for licensing and operation
- Past efforts have focused on systems using sparging gas (such as MSRE)
  - Recent efforts focus on formation of pure gases without sparging gas

MSRE core

SAM Capabilities

- Flow models (single-phase, 1-D, multi-D, …)
- Heat transfer (convective, conduction, thermal radiation, …)
- Reactor kinetics (point kinetics, decay heat, reactivity feedbacks)
- Special components (pump, heat exchanger, valves, heat pipes, steam generator, …)
- Special processes (mixing, thermal stratification, …)
- Species transport (in fluid, solid, fluid-solid and liquid-gas interfaces)
- Special models (fluid solidification, tritium transport, structure expansion, materials models…)
- Control & trip systems
- Multi-scale multi-physics coupling

- "System computer codes must be able not only to simulate several subsystems, many components and their couplings, but also the simultaneous occurrence of various phenomena and processes."

Example of species system modeled by SAM

- Species can exist in the gas phase, transported by gas velocity
- Phase transfer using Henry's Law for high-volatile gas
  - Henry and mass transfer coefficient calculated by Mole
  - Interfacial area and gas velocity by SAM
- Capture decay between species

$$\frac{\partial \alpha \rho_g}{\partial t} + \frac{\partial \alpha \rho_g v_g}{\partial x} = S$$

Gas transport equation plus drift-flux provides gas void and velocity

$$\frac{\partial I_l}{\partial t} + \frac{\partial (v_l I_l)}{\partial x} = -\lambda_I I_l + \gamma_I \bar{\Sigma}_f \Phi_f$$

Fission source from Griffin

Track decay from one species to another

$$\frac{\partial Xe_l}{\partial t} + \frac{\partial (v_l Xe_l)}{\partial x} = \lambda_I I_l - \lambda_{Xe} Xe_l - \underbrace{K_l A_i (Xe_g HRT - Xe_l)}$$

Added phase transfer term

Species can exist in gas phase

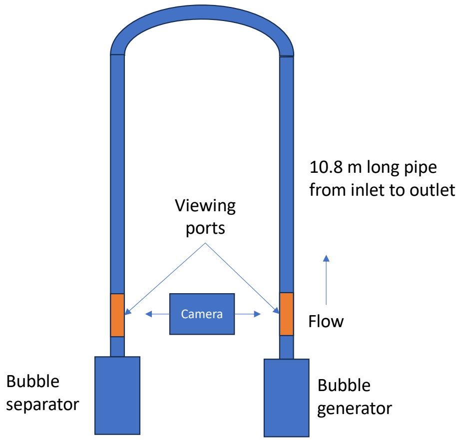
$$\frac{\partial Xe_g}{\partial t} + \frac{\partial (v_g Xe_g)}{\partial x} = -\lambda_{Xe} Xe_g + K_l A_i (Xe_g HRT - Xe_l)$$

Bubble behavior from SAM (in addition to species transport)

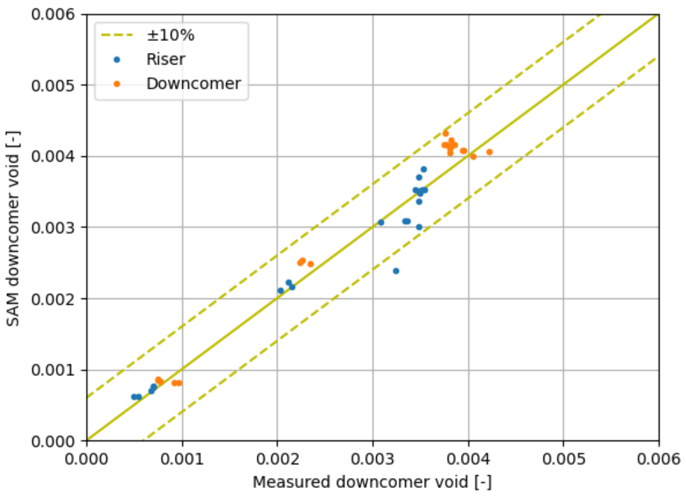
Gas model verification and validation

Model terms	Mass transient (III.A)	Momentum transient (III.B)	Energy transient (III.C)	Momentum advection (III.D)	Energy advection (III.E)	Drift flux tube (III.F)
Transient mixture mass	x					
Transient gas mass	x					
Transient mixture momentum		x				
Transient mixture energy			x			
Mixture mass advection	x	x		x		
Gas mass advection	x	x		x		
Mixture momentum advection		x		x		
Mixture energy advection				x	x	
Gas drift flux						x

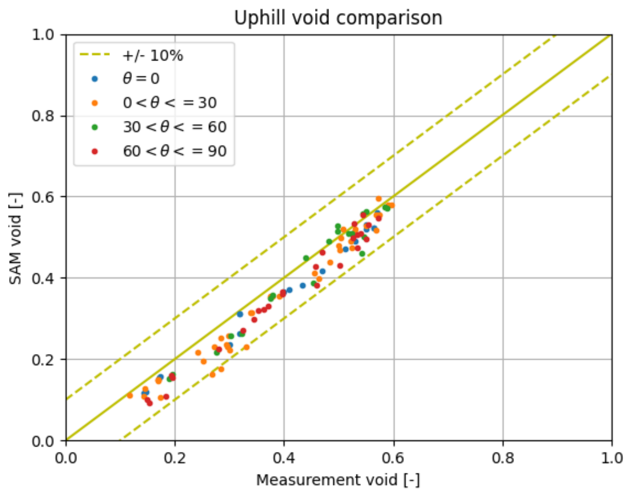
Code verification performed for a series of two-phase problems[1]



Kress[2] facility



SAM prediction of void in Kress

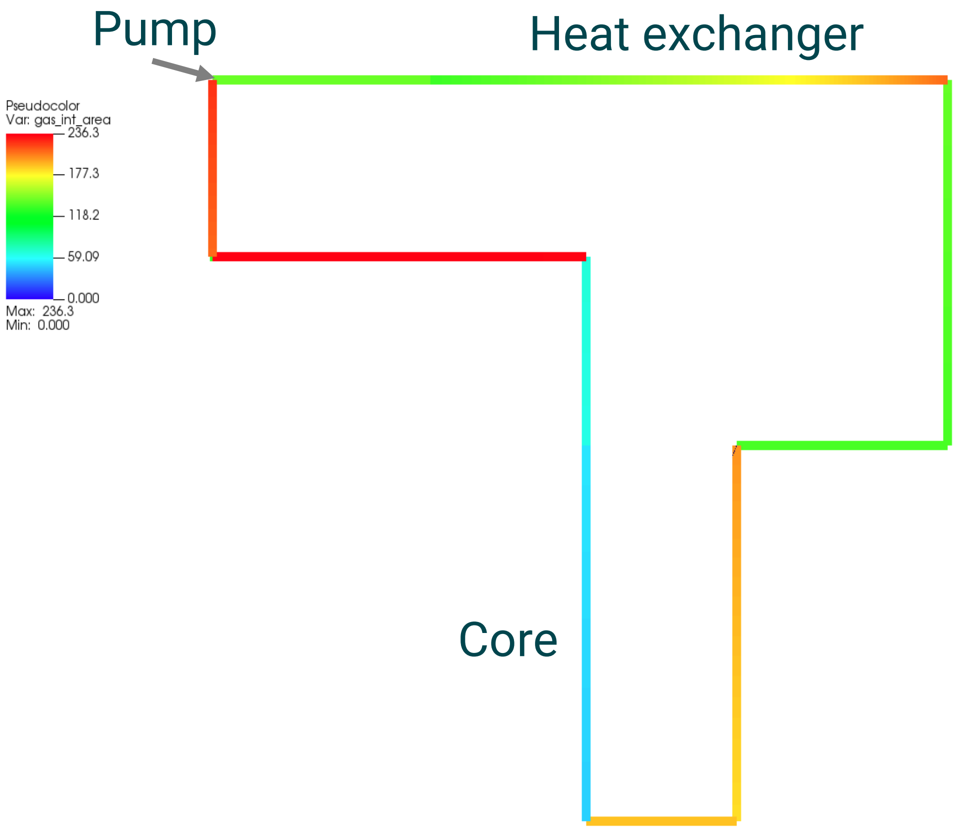


SAM prediction of void in air/water tests performed in tubes with various angles of inclination[3]

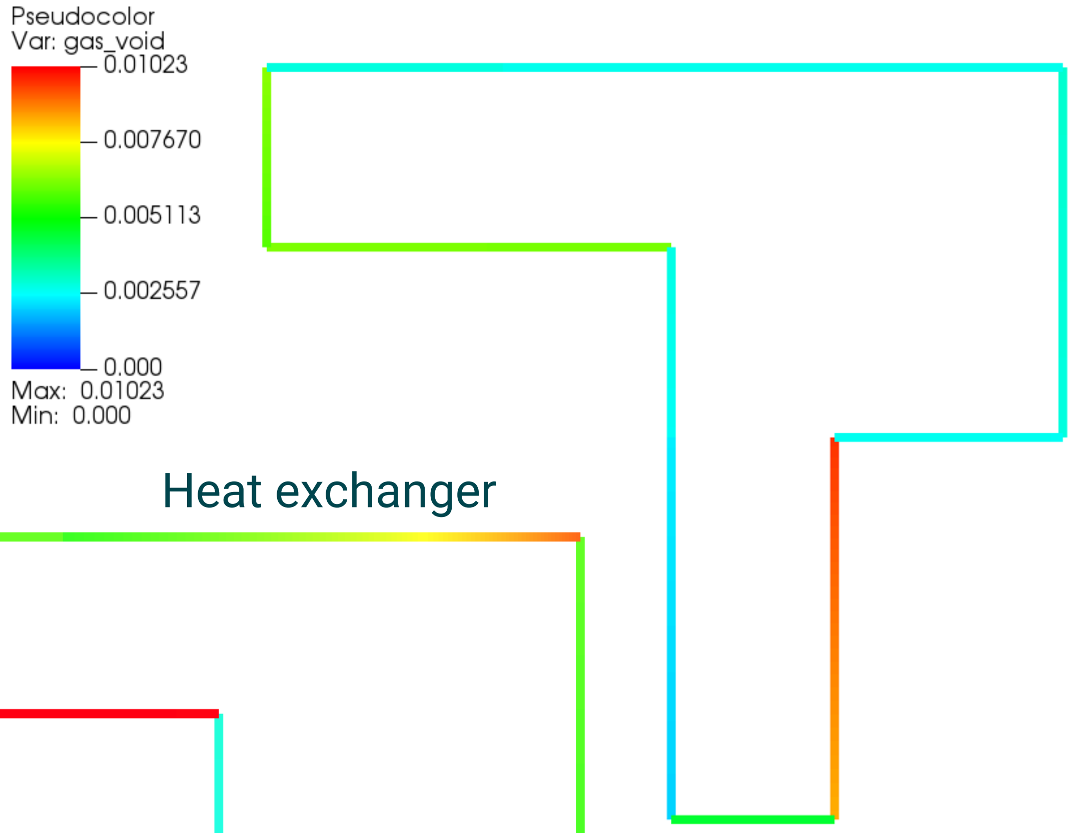
1. Salko, R, et al., “Implementation of a drift flux model into SAM with development of a verification and validation testing suite for modeling of noncondensable gas mixtures”, submitted to Nuclear Technology, 2024.
2. T. Kress, “Mass Transfer Between Small Bubbles and Liquids in Cocurrent Turbulent Pipeline Flow,” ORNL-TM-3718, 1972.
3. H. Beggs, “An experimental Study of Two-Phase Flow in Inclined Pipes,” PhD Thesis, The University of Tulsa, 1972.

Modeling of MSRE

- Sparging gas utilized in MSRE for removal of fission products
- Modeling with SAM allows for prediction of void and interfacial area distribution
- Predictions can be utilized in mass transport solution



Interfacial area distribution



Void distribution

Questions!

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